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Initial Design Calculations for a Detection System that will Observe Resonant Excitation of the 680 keV state in 238U

J. Pruet, C. Hagmann

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ABSTRACT

We present calculations and design considerations for a detection system that could be used to observe nuclear resonance fluorescence in ^{238}U. This is intended as part of an experiment in which a nearly monochromatic beam of light incident on a thin foil of natural uranium resonantly populates the state at 680 keV in ^{238}U. The beam of light is generated via Compton upscattering of laser light incident on a beam of relativistic electrons. This light source has excellent energy and angular resolution. In the current design study we suppose photons emitted following de-excitation of excited nuclei to be observed by a segmented array of BGO crystals. Monte Carlo calculations are used to inform estimates for the design and performance of this detector system. We find that each detector in this array should be shielded by about 2 cm of lead. The signal to background ratio for each of the BGO crystals is larger than ten. The probability that a single detector observes a resonant photon during a single pulse of the light source is near unity.

1. Introduction

At LLNL there is currently a concerted effort to demonstrate the utility of laser/linac-based light sources for detecting small amounts of specific isotopes in well shielded environments (Barty & Hartemann 2004). Detailed initial calculations show that interrogation systems built around these sources should be characterized by remarkably good performance (Pruet et al. 2006). Of particular interest to homeland security applications is the prediction that millimeter sized samples of the fissile weaponizable isotopes can be quickly detected even when hidden in packed cargo containers. And, the error rates (both false positive and false negative) associated with cargo interrogation are calculated to be uniquely low.

Initial efforts toward developing a working proto-type of this system are advancing on two fronts. One involves building the laser, photo-gun, optics and accelerator components that comprise the light source. The other involves collecting needed nuclear data and demonstrating that the light source works as predicted to excite strong resonances in nuclei.

Experimental campaigns aimed at finding strong electromagnetic resonances in ^{239}Pu and ^{235}U have recently been completed (Johnson et al. 2007). Usefully strong resonances were discovered. Here we report on initial calculations for the design and performance of a detection system that could be used in demonstration experiments with the new light source. These experiments involve resonant excitation of the state at 680.11 keV in ^{238}U. We chose this experiment for a few reasons. First, the 680 keV state is connected to the ground state of uranium through a rather strong M1 resonance. There is also some security interest in being able to observe ^{238}U, though not as much as in being able to detect ^{239}Pu or ^{235}U. Lastly, the fairly low excitation energy of this state makes it accessible with modest linac energies.

2. Basic Considerations for the Detection System

Figure 1 gives an overview of the experimental setup. In this setup a beam of light is incident on a thin foil of natural uranium. Interactions between the beam and the foil cause photons to be emitted at large angles relative to the incident beam. Some of these photons come from de-excitation of resonantly excited uranium atoms. This is the signal we want to detect. Some of the photons come

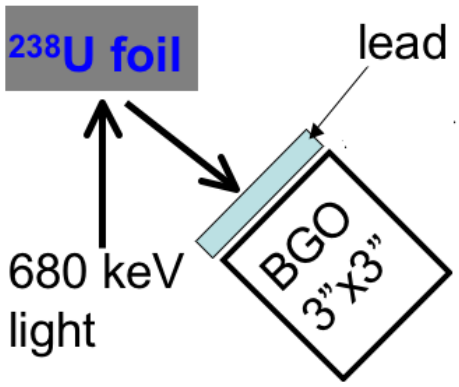

Fig. 1.— Illustration of the experimental setup for detecting NRF in ^{238}U. Here the 680 keV light is produced by a laser/linac source (Barty & Hartemann 2004) and is expected to have an energy resolution of about 1 percent. Two types of photons result from the interaction of incident light with the uranium foil. NRF photons are produced as resonantly excited nuclei decay. This process produces photons with energies of 680 keV and 635 keV. Background photons come from interactions between incident photons, electrons and atoms in the sample foil. A lead attenuator is used to filter out these unwanted photons. A segmented array of BGO detectors observes photons that survive the passage through the lead.

from other processes and just contribute unwanted background. A large portion of this background is comprised of low energy photons. To filter this out a piece of lead is placed between the foil and the detector. Since lead preferentially attenuates low energy photons it can be used to lower backgrounds while still preserving a good fraction of the wanted signal.

2.1. Characteristics of the Light Source

2.1.1. Energy Resolution

This beam has some photons within the energy window capable of resonantly exciting the 680 keV state in ^{238}U and some photons outside of this window. The width of the window is determined by thermal motion of the uranium atoms and is approximately (Metzger 1956)

$$\Gamma_{\rm thermal} = 0.34 \text{ eV}. \quad (1)$$

The width of the incident beam is denoted by $\Gamma_{\rm beam}$. Calculations show that the energy resolution of the beam should be in the sub-percent range, which implies

$$\Gamma_{\rm beam} \lesssim 10\,\text{keV}. \quad (2)$$

This is very small compared to the resolution characterizing Bremsstrahlung beams, but is still large compared to the width in eq. 1 characterizing nuclear resonance fluorescence (NRF).

2.1.2. Temporal Characteristics

Temporal characteristics of the light source place demanding constraints on the detection system. This is because the light source does not emit continuously but instead puts out pulses with a duration of about one picosecond. Feasible detectors cannot resolve signals this short - all photons coming within a picosecond window will be summed together. Since this summing confuses identification of resonant photons the system must be designed so that each detector only observes one or a few photons during each light source pulse.

2.1.3. Intensity

First estimates suggest that the light source used in the experiments will produce about 10^9 photons per pulse. With an energy resolution of

1% and the NRF resonance width given above, this implies that there are about $5{\cdot}10^4$ resonant photons per pulse. This is a large number and implies that pulse pileup must be considered.

2.2. Backgrounds

Hagmann & Pruet (2006) and Pruet et al. (2006) present a detailed discussion of the backgrounds resulting from interactions between the incident beam and the target foil. Figure 2 shows the spectrum of photons produced at angles larger than 90 degrees relative to the incident beam. Results are shown for background spectra that have passed through different thicknesses of lead. Note that as the lead attenuator becomes thicker the low energy portion of the backgrounds becomes less and less important.

Three main processes give rise to calculated backgrounds:

- Compton Scattering

  Compton interactions with electrons in the target foil dominate production of photons at large angles relative to the incident beam. Fortunately these have low energies and are easily shielded out. A 680 keV photon scattered through 90 degrees is left with an energy of 292 keV. A 680 keV photon scattered through 180 degrees is left with an energy of 186 keV.

- Elastic Scattering

  Several processes contribute to large angle elastic scattering of photons from the target foil. For photons with an energy of 680 keV Rayleigh scattering dominates. This process is included in our calculations.

- Multi-Step processes

  Hagmann & Pruet (2006) present a detailed discussion of multi-step processes that dominate production of backgrounds at energies higher than can be reached through Compton scattering. Our calculations include these processes. The term "multi-step" is used because the generation of high energy backgrounds is mediated by an electron in the target foil.

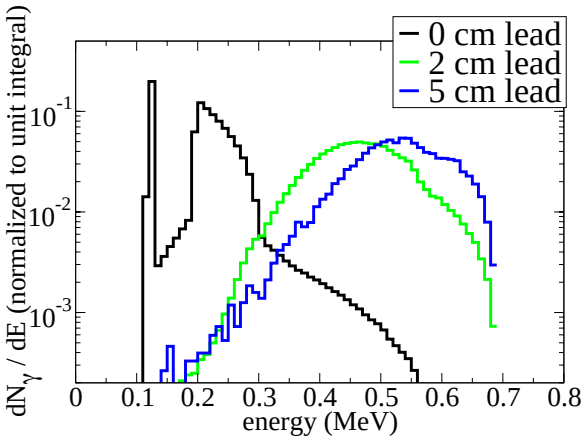

Fig. 2.— Spectrum of photons incident on a BGO detector for a beam of 680 keV light with 1% bandwidth incident on a 2 mm thick piece of uranium. The spectra here have been averaged over all backscattered directions. Different curves here correspond to spectra that have passed through different thicknesses of lead. To illustrate the dependence of the shape of the spectra on the lead thickness each spectrum has been normalized to unit integral. Note that the spectra harden as the lead becomes thicker. This arises because higher energy photons have a smaller interaction cross section with lead (for the energies shown here).

2.3. NRF characteristics of the 680 keV state

The 1^- state at an excitation energy of 680.11 keV in ^{238}U has a measured half life $\tau = 35+19-9$ fs. It decays to two states - the ground state with a branching ratio of 44% and an excited state at 44.9 keV with a branching ratio of 56% (Chukreev, Makarenko & Martin 2002). The thermally averaged cross section for a resonant photon having an energy within a window of width $\Gamma_{\rm thermal}$ centered around the resonance energy is

$$\sigma_{\rm NRF} \approx 200 \pm 70 \text{ barns.} \quad (3)$$

To a good approximation photons emission during de-excitation of the 680 keV state is isotropic. It is worth noting that the NRF cross section is much larger than the cross section for atomic interactions at these energies

$$\sigma_{\rm atomic} \approx 46 \,\text{barns.} \quad (4)$$

3. Detection System Design

3.1. Design Goals

For this detection system we assume that the basic goal is to positively observe resonance fluorescence in the target foil as quickly as possible. There are other possible goals. For example, one might want to measure the cross section for resonant excitation of the 680 keV state as accurately as possible. The design of a system with that goal would be quite different than the one described here.

3.2. Choice of Detectors

There are two requirements for achieving quick, confident detection of resonance fluorescence. The first involves getting a high efficiency for detecting NRF photons. The second involves reducing backgrounds that interfere with detection.

We have found that 3 in $\times$ 3 in BGO detectors satisfy both requirements. High efficiency is simply a result of the excellent intrinsic efficiency - the full peak efficiency is approximately 90% at 680 keV - characterizing these crystals. In section 3.4 we present calculations showing that backgrounds recorded by these detectors are small.

3.3. Thickness of the Uranium Foil

The thickness of the uranium foil can be chosen to minimize the needed detection time. There are two considerations here. The first is just that the efficiency for producing a resonant photon should be large. This sets a lower limit on the thickness of the foil. The second consideration relates to the backgrounds that interfere with detection. These increase with increasing foil thickness and so set an upper bound on the thickness of the foil.

To understand the constraint on foil thickness implied by the demand for high efficiency note that the mean free path of a photon in natural density uranium is

$$\lambda = 22.8\,\text{cm}\left(\frac{1\,\text{barn}}{\sigma_\gamma}\right). \quad (5)$$

Here σ_γ is the total interaction cross section for the photons. For resonant photons this is comprised of an atomic piece and the nuclear absorption piece: $\sigma_\gamma = \sigma_{\rm NRF} + \sigma_{\rm atomic} \approx 250$ barns. This implies a mean free path

$$\lambda_{\rm res} \approx 0.80\,\text{mm.} \quad (6)$$

The efficiency for producing NRF photons saturates once the foil thickness is larger than about $\lambda_{\rm res}$.

The size of backgrounds from multi-step processes and Compton scattering scales approximately linearly with foil thickness for foil thicknesses near $\lambda_{\rm res}$. To see if a careful tuning of the foil thickness is warranted we show in figure 3 the signal expected on a BGO detector for a 2 mm thick foil and 2cm of lead shielding between the foil and the detector. At this foil thickness the size of the NRF signal is saturated. And, it is seen from the figure that the signal to noise ratio is larger than 14. Because this is already fairly good we simply choose a foil thickness of 2 mm.

3.4. The thickness of lead shielding and the spectrum seen by the BGO

As we noted above, the duration of each pulse of the light source is much smaller than the temporal resolution - or smallest time separating distinguishable events - characterizing our detectors. With spectroscopy the best one can hope for is to collect a single NRF photon in each detector

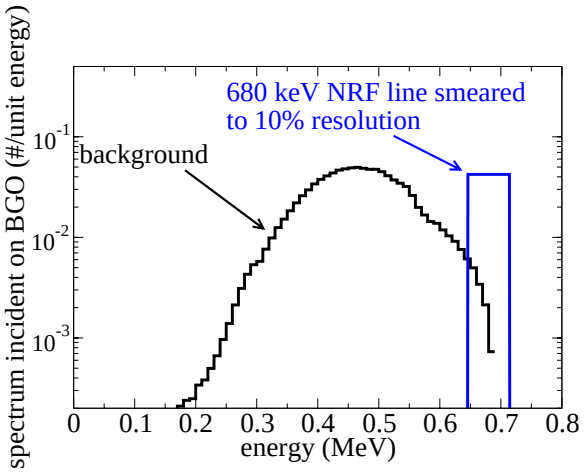

Fig. 3.— Spectrum of photons incident on a BGO detector for a beam of 680 keV light with 1% bandwidth incident on a 2 mm thick piece of uranium. The spectrum shown has been averaged over all back-scattered directions. The attenuator between the foil and detector is comprised of a 2 cm thick piece of lead. The blue box here represents the spectrum of the 680 keV line resulting from de-excitation of excited ^{238}U and smeared out over a 10% energy width. This is approximately the resolution of BGO detectors. The ratio of the number of NRF counts in this box to the number of background counts is about 14.

during each pulse. If the backgrounds were low enough one could also use simple dosimetry or gross counting instead of spectroscopy to detect NRF. This simpler approach is possible for the detection of the light elements that are characterized by small backgrounds. Dosimetry is also feasible for the detection of actinides and other high Z elements provided that the energy resolution of the light source is fine enough. For example, we will see below that if the resolution of the light source were 0.1% (rather than the 1% assumed here) the NRF resonance in ^{238}U could be readily observed with simple dosimetry. This simplifies the detection system enormously. For the present, though, we will concentrate on spectroscopic identification of NRF.

There are two different ways to design a spectroscopic detection system. One corresponds to the limit where the light source is not very bright, while the other corresponds to a bright light source. With a dim source, one can't hope to observe a single NRF photon every time the light source is fired. There just aren't enough photons. In this case one would design the system to maximize the number of photons collected by the detector. With a bright light source the problem is different, there are more photons than can be used. In this case one tries to design the system to (i) collect only one photon per pulse and (ii) maximize the probability that this photon is an NRF photon and not a background photon.

To see if our planned experiments correspond to a bright source or a dim source we note that the light source used in the experiments is expected to produce more than 10^4 resonant photons per pulse (see section 2.1.3). The foil thickness was chosen so that the efficiency for an incident resonant photon to produce NRF is of order unity. Our BGO detectors have an absolute efficiency (including the effect of the solid angle) for detecting photons emitted by the foil of order 0.001. Put together these numbers imply that our light source is bright enough to produce more than 10 NRF counts in each detector for each pulse.

A lead attenuator placed between the foil and detector will be used to prevent pulse pileup in the detectors. We could also prevent pulse pileup by dimming the light source or moving the detectors further from the foil, but this would result in unwanted backgrounds making up most of the

observed signal. High Z materials like lead preferentially absorb the unwanted backgrounds.

To determine the optimum lead thickness we show in table 1 the signals expected for different thicknesses of lead shielding. Results for both the NRF photons and the background photons are shown. It is seen that the ratio of NRF photons that survive passage through the lead to background photons that survive this passage is very small if no lead shielding is used. This ratio approaches unity for a 2 cm thick piece of lead. If we assume that BGO detectors are placed 20 cm from the foil, then the number of photons collected per detector per pulse is also about unity. This suggests that approximately 2 cm of lead shielding is appropriate. If more shielding is used the NRF/background ratio gets better, but the absolute efficiency for detecting a photons becomes too small.

Figure 3 shows the spectrum of photons incident on the BGO for 2 cm of lead shielding. The spectrum shown has been averaged over all backscattered directions. The NRF signal in this plot has been smeared over an energy window of width 10 %, which is approximately the energy resolution of BGO detectors. The ratio of the number of NRF counts to background counts in this energy window is larger than 10. This implies easy spectroscopic discrimination.

4. Summary

We've presented initial calculations describing the performance and design of a segmented array of BGO detectors in an experiment aimed at observing resonance fluorescence in ^{238}U. It is assumed that the experiment will be driven by a light source that has an energy resolution of about 1% and that emits approximately 10^9 photons per pulse. In this case the uranium foil should be one or two mm thick. Each BGO detector should be shielded from direct sight of the foil by about 2 cm of lead and be separated from the foil by a distance of about 20 cm. With this configuration each BGO detector has about a 50% chance of observing a resonant photon per pulse and problems associated with pulse pileup are ameliorated. For a single BGO detector and a 10 Hz light source, confident detection of NRF in the uranium foil takes about 20 seconds. Needed observation time scales inversely with the number of detectors.

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TABLE 1

BACKGROUND AND NRF COUNTS FOR DIFFERENT ATTENUATOR THICKNESSES[a]

Lead Thickness (cm)	NRF Photons[b]	Background Photons[c]	ratio (NRF/Background)
0	$4.0 \cdot 10^{-5}$	$1.2 \cdot 10^{-2}$	0.03
1	$1.3 \cdot 10^{-5}$	$6.3 \cdot 10^{-5}$	0.25
2	$4.2 \cdot 10^{-6}$	$7.2 \cdot 10^{-6}$	0.6
5	$1.4 \cdot 10^{-7}$	$5.4 \cdot 10^{-8}$	2.6

[a]Here the beam is assumed to have an energy resolution of 1%. Background scales linearly with beam resolution for small beam resolution. The attenuator between the uranium foil and BGO detector is lead. All numbers here are normalized to unit incident photon on the uranium foil.

[b]This includes both the line from decay of the 680 keV state to the ground state and the line from decay of the 680 keV state to the state at an excitation energy of 45 keV in ^{238}U.

[c]This includes photons of all energies, not just those near 680 keV.